

LA-UR-18-30579

Approved for public release; distribution is unlimited.

Title: Measuring Monodisperse Aerosol Transmission in the Los Alamos
Respirable Release Fraction Measurement Chamber

Author(s): Tao, Yong
Moore, Murray E.

Intended for: Report

Issued: 2018-11-02

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Measuring Monodisperse Aerosol Transmission
in the Los Alamos Respirable Release Fraction Measurement Chamber

Yong Tao and Murray E. Moore
Radiation Protection Services, Aerosol Engineering Facility
Los Alamos National Laboratory
Los Alamos, NM 87544

ABSTRACT

The transmission ratio of monodisperse aerosol (between 1 and 20 μm aerodynamic equivalent diameter) was measured in a modified configuration of the Los Alamos RRFMC (Respirable Release Fraction Measurement Chamber). The RRFMC is a newly built, 20-foot tall, airtight chamber for the drop testing of nuclear material storage containers. The RRFMC was integrated into an existing HEPA filtered wind tunnel system, and is capable of direct measurement of Airborne Release Fraction (ARF) and Respirable Release Fraction (RRF) for plutonium and uranium under various accident conditions. Cerium oxide powder is used as a surrogate for plutonium oxide. Real-time aerosol measurements for aerodynamic diameters from 0.5 to 20 μm are performed with a TSI Inc (Shoreview, MN) Model 3321 APS (Aerodynamic Particle Sizer) spectrometer.

Containers are dropped in a vertical tower, and HEPA filtered air carries the released powder through a mixer system to the aerosol spectrometer. The mixer contains components developed at both Los Alamos and Texas A&M University, and this composite mixer establishes conditions suitable for using a single nozzle on the aerosol spectrometer. This single point capability is in accordance with relevant portions from ANSI N13.1-2011 (Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities). The downstream sampling location therefore moves the aerosol counter away from the high velocity impact test zone, and aerosol mixing occurs in a previously validated duct system. Deposition losses during the transport of test powder from the impact area to the air sampling location were determined (US EPA 40 CFR 53.42). This method used monodisperse oil droplets tagged with sodium fluorescein tracer to measure aerosol transmission by simultaneous sampling of reference and test particle concentrations. Based on the experimental results, the transmission ratio for the aerodynamic diameters for every APS sampling channel was calculated. These estimated transmission ratio values then produce experimentally estimated values for the ARF and RRF for container drop testing.

Keywords: Aerosol, Transmission, Air sampling, Respirable Release Fraction

INTRODUCTION

The airborne release of radioactive material from nonreactor nuclear facilities has been a subject of investigation for almost six decades. The publication “Airborne Release Fraction/Rate and Respirable Fractions for Nonreactor Nuclear Facilities” (DOE-HDBK-3010-94) is a well-documented handbook and it is widely used at DOE sites. The “3010 handbook” provides values derived from assessing previous experimental data that may be used in safety analyses when the

data are applicable. However, previous experimental situations may not be representative of the actual full-scale situation.

The Respirable Release Fraction Measurement Chamber (RRFMC) at the Aerosol Engineering Building, Los Alamos National Laboratory (LANL) directly measures airborne and respirable release for the drop testing of nuclear material storage containers. The RRFMC is an airtight 20-foot tall PermaCon™ enclosure (Pajarito Scientific Inc, Santa Fe, NM) integrated into a closed loop wind tunnel. The tunnel has HEPA filtration upstream and downstream of the drop test impact zone (Figure 1), and has been in operation since early 2005.

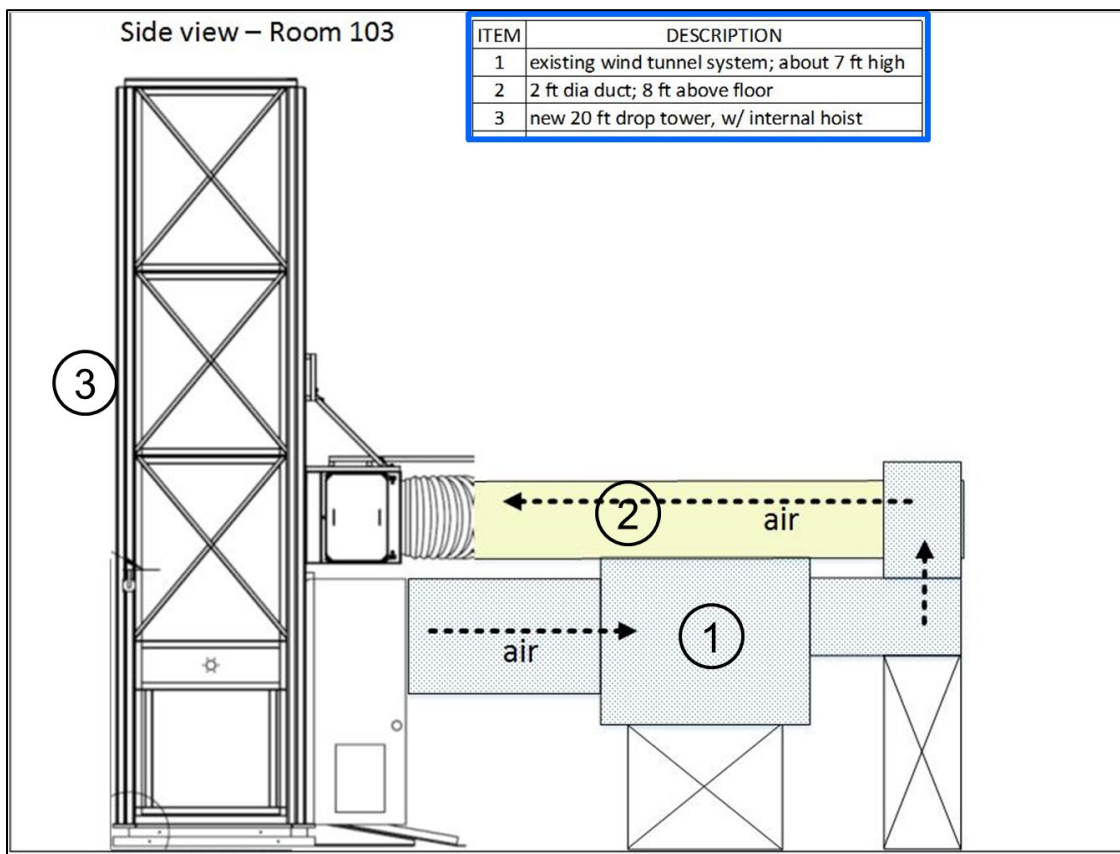


Figure 1. Side View of Filtered wind tunnel with RRFMC.

A drop test machine (Model PDT 80, Lansmont Inc, Monterey, CA) is located inside the PermaCon™ enclosure (Figure 2a). The drop test machine features a remote controlled electric hoist to raise and lower the drop leaf assembly and provide variable drop heights. It can perform drop tests on items weighing up to 150 lbs and drop heights up to 16 feet. Two MEMRECAM HX camera systems are installed for dual-axis high-speed videos and pictures (Figure 2b). For this work, cerium oxide (CeO_2) powder was selected as a surrogate for plutonium oxide (PuO_2) in release fraction experiments. Several US national labs have published articles about CeO_2 as a PuO_2 surrogate (Moore and Tao 2017).

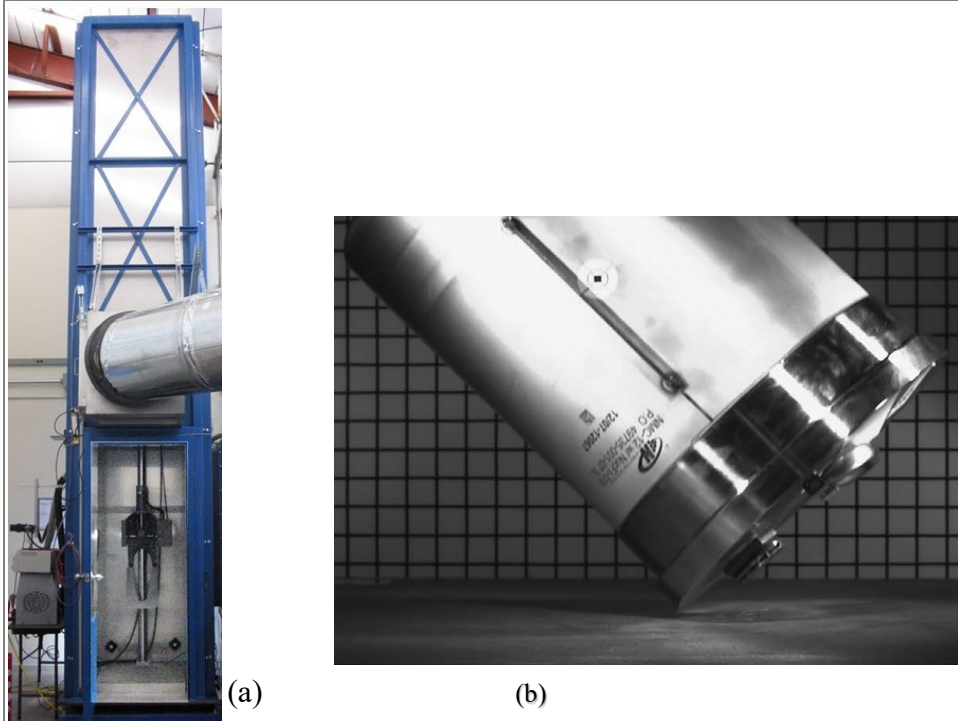


Figure 2. (a) Lansmont PDT 80 drop tester inside 20 feet RRFMC PermaCon™ enclosure, (b) Still photo of a container just before impact in a drop test.

Referring to the schematic in Figure 3, containers are dropped in the RRFMC while the wind tunnel is actively filtering the airflow. If any test powder is released from a dropped container at the impact #1, the powder is swept into a “generic mixer” #3 (McFarland et al. 1999) and into a duct #4 for single point sampling (ANSI 2011 and Moore et al 2010). Real-time aerosol measurements for aerodynamic diameters from 0.5 to 20 μm are performed with an APS at location #6. The test air is filtered in a bank of HEPA filters #8, into a fan #11, through an overhead duct #12, then through another HEPA filter #13. The airflow is routed through a plenum #14 that then sweeps an air curtain of purified air across the impact zone surface #1.

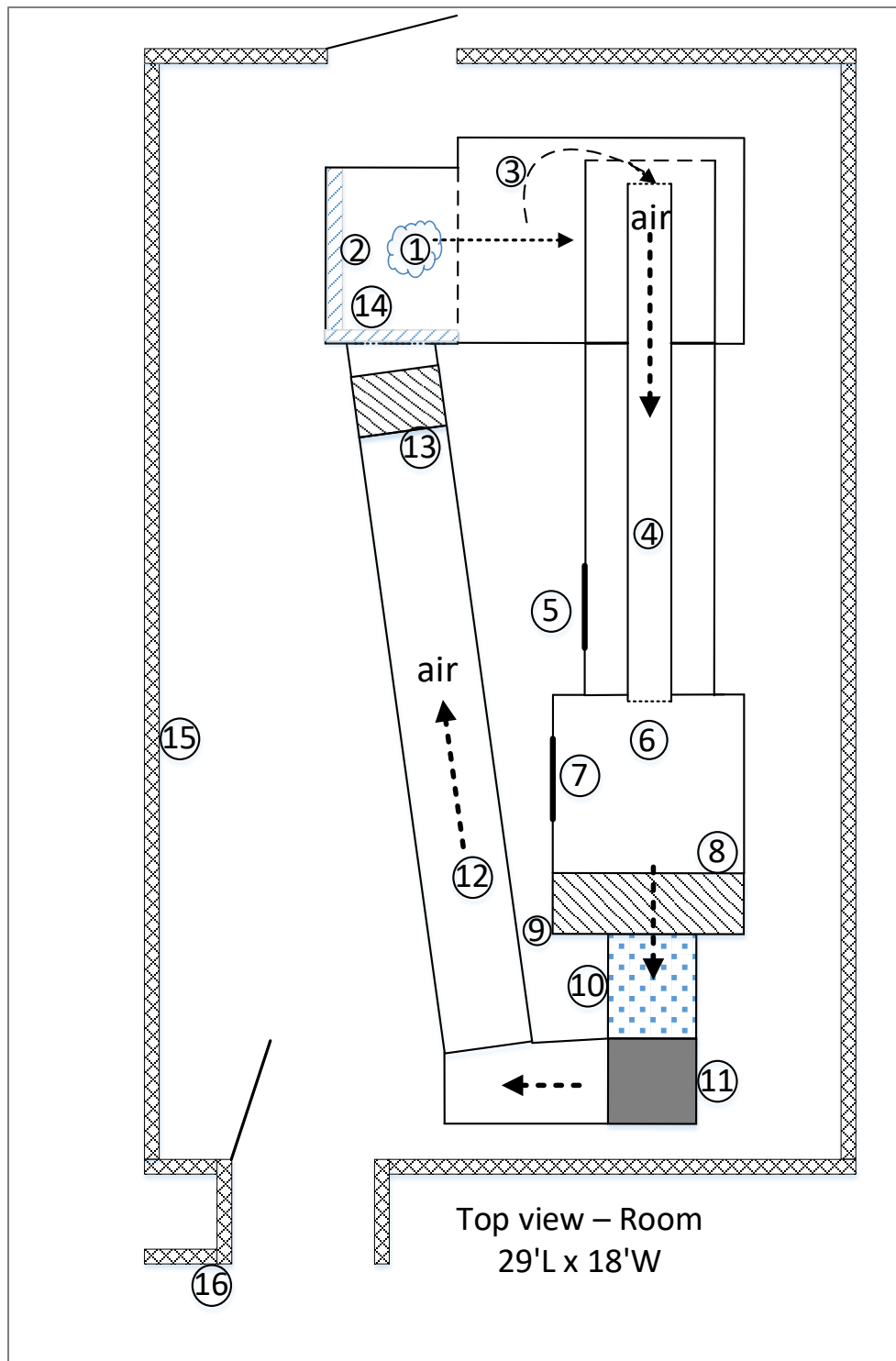


Figure 3. Top view of the schematic floor plan and wind tunnel with RRFMC.

Table 1. Legend for Figure 3 and equipment status of aerosol transmission test.

RRFMC air flow schematic - 9-13-2018.xlsx	
ITEM	DESCRIPTION
1	Drop impact location, with routing plenum in tower section
2	Pressure tap location for the drop tower
3	Air mixer - Apply silicone spray on internal surfaces to reduce aerosol resuspension upon load impacts. (HEPA filters removed.)
4	Duct (Dia=1 ft) with velocity probe
5	Hatch to HEPA filter in the duct (removed for drop testing)
6	Aerodynamic particle sizer (APS) in wind tunnel test section
7	Hatch to wind tunnel (closed)
8	HEPA filters at test section outlet (Qty=4 in series)
9	One hatch to HEPAs open, gapped with 0.25 inch spacers
10	6-speed switch for wind tunnel speed control
11	Fan for wind tunnel
12	Overhead duct (Dia=2 ft)
13	HEPA filter at drop tower inlet (Qty=1)
14	Plenum to route air flow inside the drop tower
15	Lockout for drop tower
16	Drop tower and wind tunnel remote control keypads
<p>Test type: <u>Drop Test</u></p> <p>Blower speed: 24.0 Hz to produce about 1.4 m sec^{-1} at location #4.</p> <p>RRFMC tower $\Delta P(\text{inWC})$: 0.05 inWC</p>	

MATERIALS AND METHODS

The transmission ratio of aerosol from the impact area #1 to the measurement location #6 was measured with monodisperse fluorescent oil tracer particles in the RRFMC (Tao and Moore 2018). Previous aerosol measurements in this tunnel (Moore et al 2007) were consulted. An aerosol injection duct was built inside drop tower (location #1, Figure 5). Aerosols were generated by a VOMAG (Vibrating Orifice Monodisperse Aerosol Generator) (TSI Inc, Model 3450). The particle size and distribution of monodisperse aerosol was measured by the APS. Aerosol was injected 5 feet above (Figure 4, 7, and 8) the impact zone #1 and mixed with flow (around 0.40 m/s) from the air curtain (created by the plenum). The test aerosol entered the impact zone #1 through the aerosol injection duct (cross section $0.155 \text{ m} \times 0.085 \text{ m}$). A glass fiber filter in an open-face holder faced the end of the injection duct for reference sampling (Figure 5). An identical test filter was at the end of the 6 feet long (1 foot diameter) duct #4, pointed into the airflow (Figure 6). Flow meters for the reference filter Q_{ref} and test filter Q_{test} (TSI Model 4045 and AliCAT Model 13M-100SLPM-D/5M) regulated the airflow to about 30 ALPM (ambient liters per minute). The aerosol injection duct flow rate, q_{ref} , and mixing duct #4 flow rate, q_{test} , were measured with a calibrated Model 9565-P TSI velocity probe (TSI Inc., Shoreview, MN).

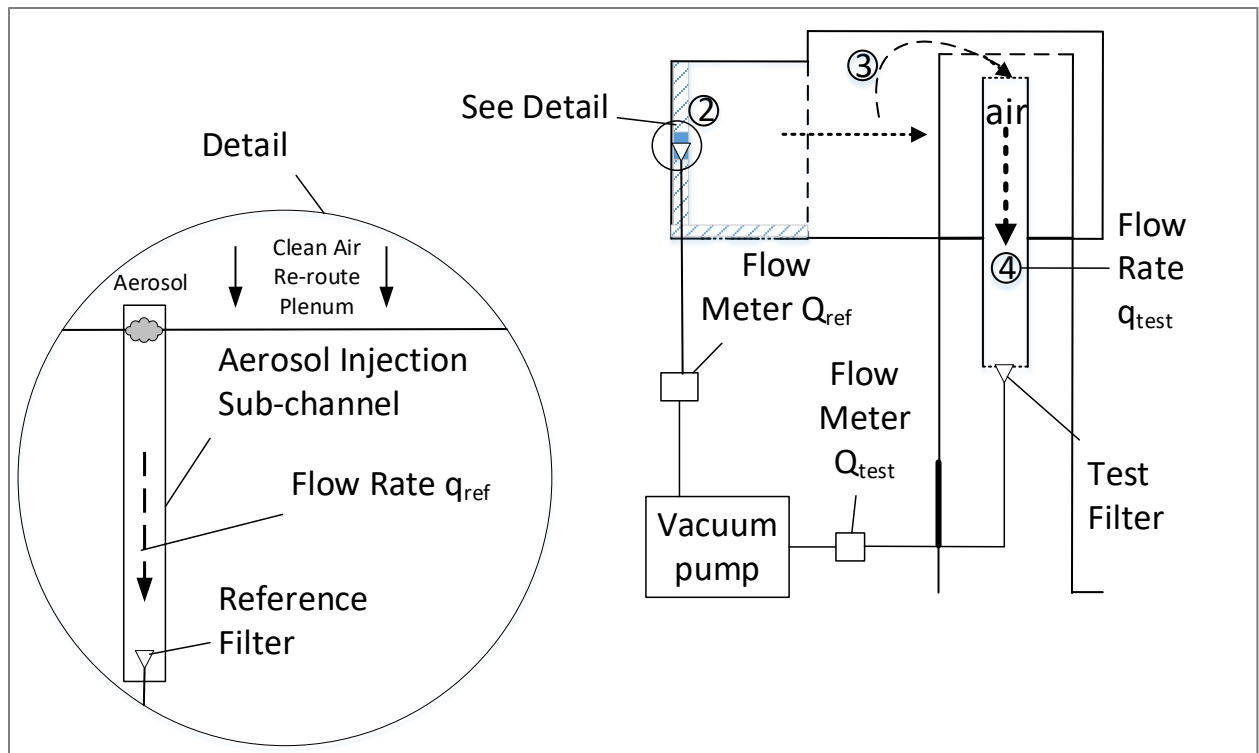


Figure 4. Schematic diagram of experimental set up for aerosol transmission test.

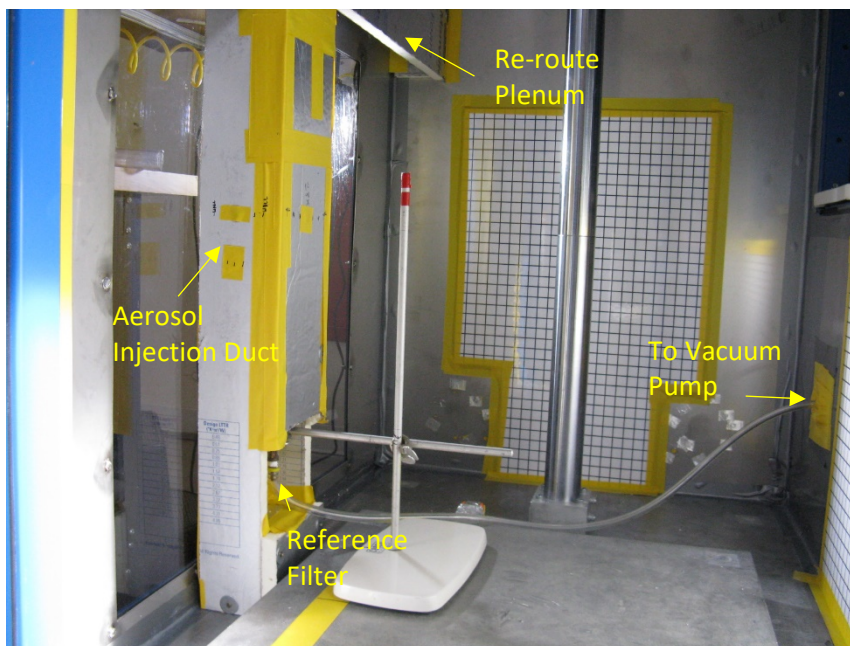


Figure 5. Reference filter located in RRFMC.

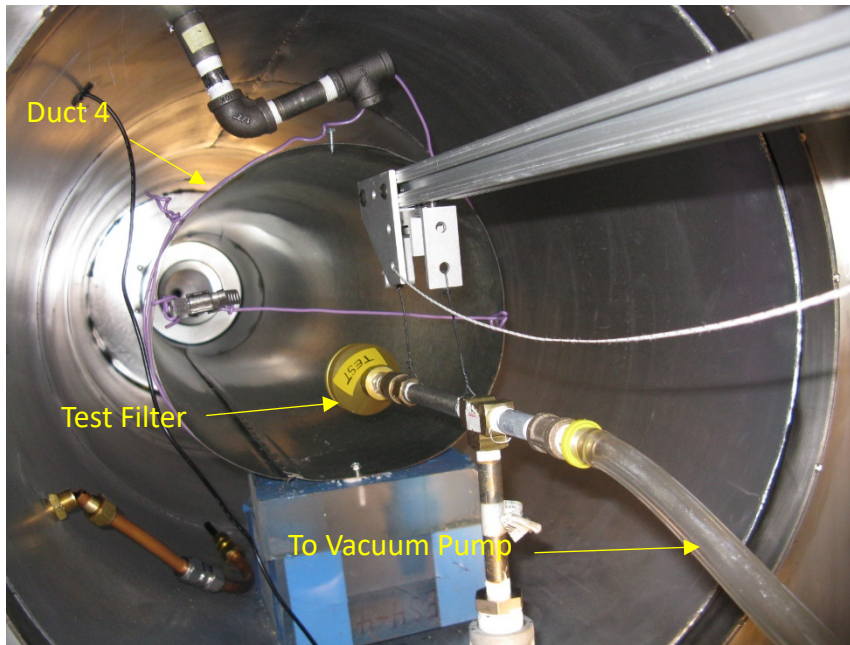


Figure 6. Test filter located at the end of duct #4.

Figure 7 shows the P&ID diagram of aerosol transmission test. Figure 8 shows the different views of aerosol injection sub-channel in RRFMC.

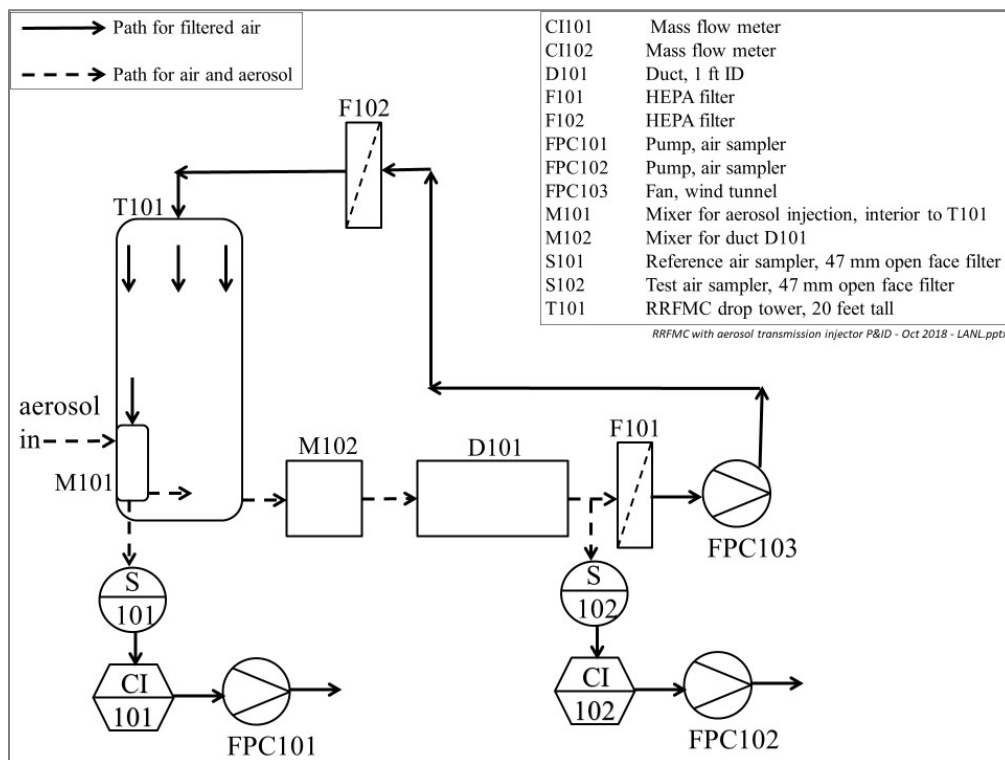


Figure 7. P&ID diagram of RRFMC aerosol transmission test.

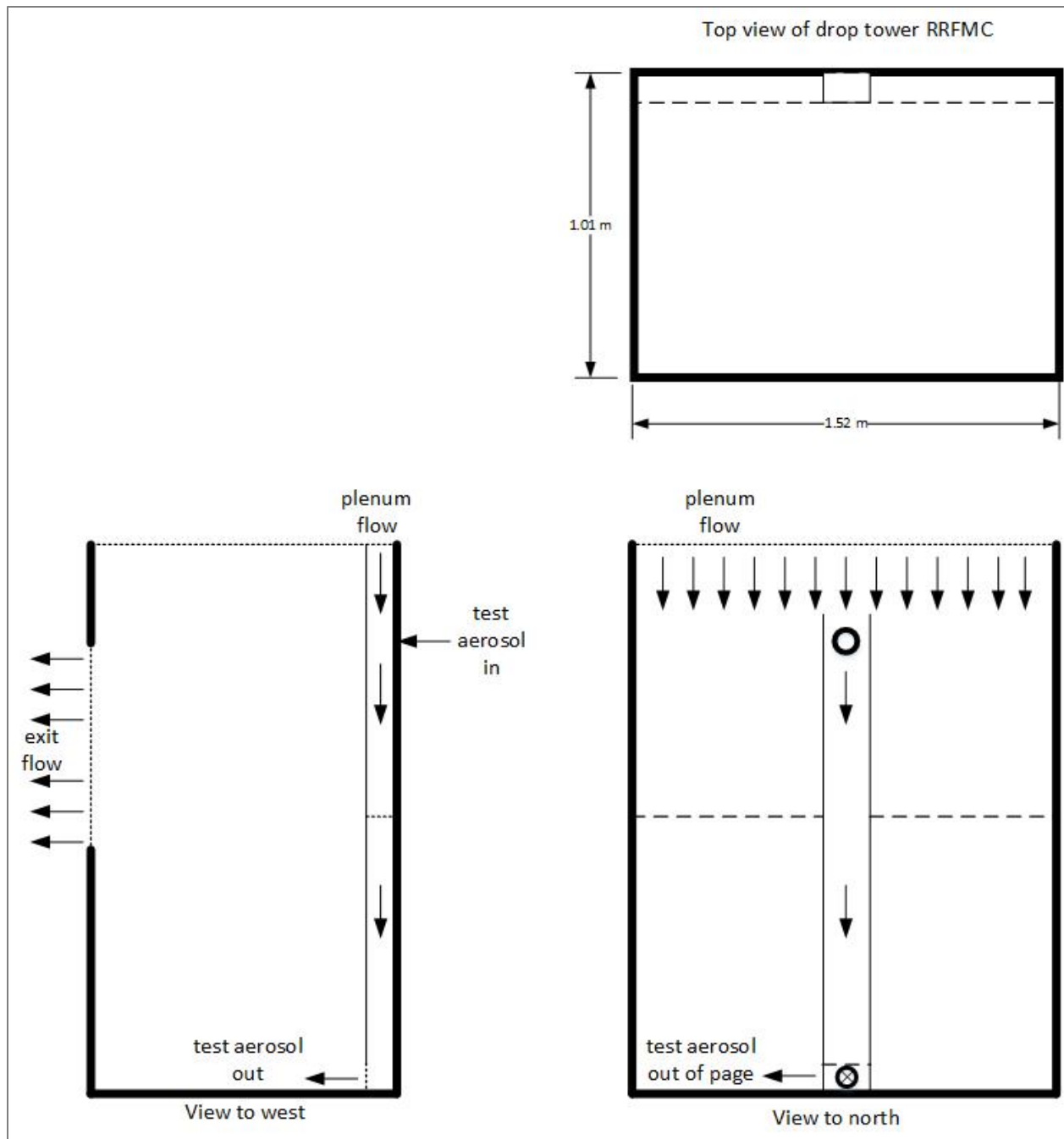


Figure 8. Sketches of aerosol injection sub-channel.

After aerosol collection, reference and test filters were placed in a 50/50 solution (aqueous NaOH, pH=8.3) and isopropyl alcohol, and the fluorescent oil aerosol was allowed to dissolve overnight. The fluorescent dye concentrations were measured on a relative basis with a fluorometer (Turner Model 450). Clean glass fiber filters in the same solution were used to zero the fluorometer. After a few seconds of hand-agitation, the liquid solutions from the beakers were sequentially poured into a glass fluorometer test cuvet. The fluorometer readings from three consecutive samples of liquid from each beaker were recorded and the average of the three readings was calculated.

The relative fluorescent concentration, C , present in each measured solution was calculated:

$$C = \frac{FL}{QT} \quad (1)$$

Where: C = relative concentration of fluorescent aerosol in the sampled air,
 F = average reading on the fluorometer of the liquid sample,
 L = liquid volume in the beaker of alcohol and water,
 Q = measured sampling air flow rate through the filter, and,
 T = sample time through the filter.

Monodisperse aerosol transmission ratio for RRFMC was quantified:

$$P = \frac{m_{measured}}{m_{injection}} = \frac{C_{test} \times q_{test}}{C_{ref} \times q_{ref}} \quad (2)$$

Where: P = the transmission ratio of tested monodisperse aerosol,
 C_{test} = the concentration from the test air sampler,
 q_{test} = the air flow rate in duct #4,
 C_{ref} = the concentration from the reference air sampler, and,
 q_{ref} = the air flow rate in aerosol injection duct.

Substituting equation (1) into equation (2),

$$P = \frac{(\frac{FL}{QT})_{test} \times q_{test}}{(\frac{FL}{QT})_{ref} \times q_{ref}} \quad (3)$$

Since the sampling time, T , and liquid volume, L , of the test and reference filters are the same, equation (3) can be simplified as:

$$P = \frac{\frac{F_{test}}{Q_{test}} \times q_{test}}{\frac{F_{ref}}{Q_{ref}} \times q_{ref}} \quad (4)$$

Where: Q_{test} = the air sampling flow rate from the test air sampler, and,
 Q_{ref} = the air sampling flow rate from the reference air sampler.

The fan speed in the wind tunnel was 24 Hz. In duct #4, this yielded an air velocity of about 1.4 m/s and the flow rate, q_{test} , was about 0.1 m³/s. In the aerosol injection duct, the air velocity was about 0.4 m/s and the flow rate, q_{ref} , was about 0.005 m³/s.

RESULTS

Three replicate tests were conducted for aerosol aerodynamic particle sizes of 3.2 μm, 11.2 μm, and 16.7 μm (GSD < 1.10) respectively. Three replicate tests for one given aerosol particle size constituted a single “set”, and generated a single data point on the graphed results (Table 2 and Figure 9).

Test uncertainty was calculated by standard methods (ASME 2005) to be 18.3%. The largest single uncertainty was the aerosol concentration variation at test location #6, which was 16.8% (Moore et al. 2010).

Table 2. Measured Transmission Ratio

Aerodynamic diameter, μm	Measured transmission ratio from impact zone to aerosol sampler location
3.2	0.872
11.2	0.669
16.7	0.376

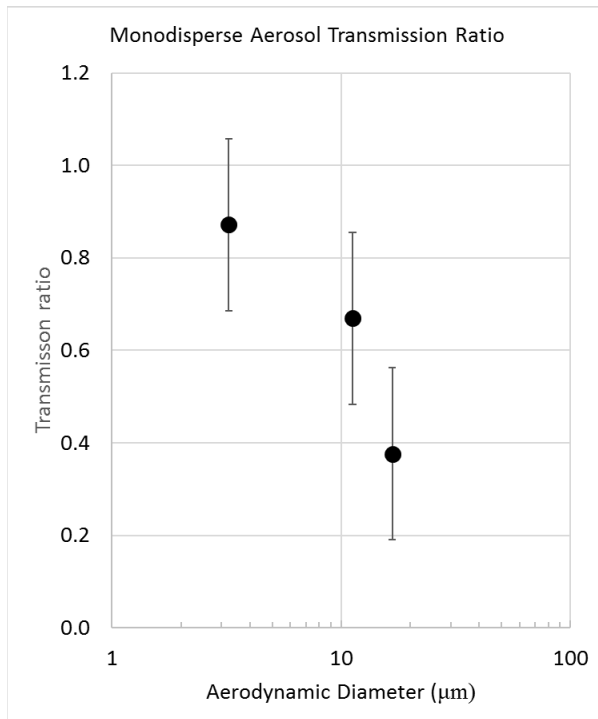


Figure 9. Monodisperse aerosol transmission ratio from impact zone to aerosol sampler location.

DISCUSSION

For aerosol transport from the impact zone to the air sampler location, the aerosol transmission mechanisms are affected by gravitational settling, diffusion, and impaction. All work in the current study and future drop tests will be performed at a constant (wind tunnel) fan speed and therefore a constant air velocity. Drag force and gas resistance effects on the aerosol transmission loss are therefore constant, and the only variable affecting the transmission ratio would be the particle aerodynamic diameter, D_a . From Stokes law (Hinds, 1999), the terminal settling velocity of a particle in air, V_{TS} , is expressed as:

$$V_{TS} = \frac{\rho_0 Da^2 g}{18\eta} \quad (5)$$

Where:

Da = particle aerodynamic diameter,
 ρ_0 = unit particle density (1.0 g cm^{-3}),
 g = gravitational acceleration, and,
 η = air viscosity.

For constant values of ρ_0 , g , and η , equation 5 can be simplified as:

$$V_{TS} = aDa^2 \quad (6)$$

where,

a = constant.

The actual case of aerosol transmission is very complicated. Firstly, consider about how aerosol concentration decays in a room or a container. For monodisperse aerosols, the real decay is closer to the stirred setting case (Hinds, 1999). The concentration of aerosol undergoing stirred settling decays exponentially with time and V_{TS} . For stirred settling:

$$\frac{n(t)}{n_0} = \exp\left(\frac{-V_{TS}t}{H}\right) \quad (7) \quad (\text{Hinds, 1999})$$

Where: n_0 is the initial aerosol concentration; $n(t)$ is the aerosol concentration at time t ; t is time; V_{TS} is particle's terminal settling velocity; H is the height of chamber.

Apply the stirred settling equation into RRFMC transport system. In RRFMC system, when $t=0$, aerosol is released at impact location; when $t=t$, the released aerosol is transported to the air sampling location #6. Since the flow rates of impact location and air sampling location changed, the aerosol concentration change does not follow equation 7. However, the total mass change should follow equation 7. Therefore, equation 7 can be rewritten for RRFMC system as:

$$\frac{m_{measured}}{m_{injection}} = \frac{m(t)}{m_0} = \exp\left(\frac{-V_{TS}t}{H}\right) \quad (8)$$

In RRFMC system, H is a constant. At constant fan speed, the time for aerosol transport from the impact zone to the sampling location, t is also constant. Therefore, equation 8 can be simplified as:

$$P = \frac{m_{measured}}{m_{injection}} = \exp(bV_{TS}) \quad (9)$$

Where: P is the transmission ratio; b is a constant, which is an operational characteristic constant of the system.

Substituting equation 5 into equation 9,

$$P = \frac{m_{measured}}{m_{injection}} = \exp(abDa^2) \quad (10)$$

Where: P is the transmission ratio; Da is the particle's aerodynamic diameter; a and b are constants.

Re-writing equation 10 as:

$$\ln(P) = (ab)Da^2 \quad (11)$$

Therefore, a plot $\ln(P)$ versus Da^2 will yield a linear relationship (Figure 10).

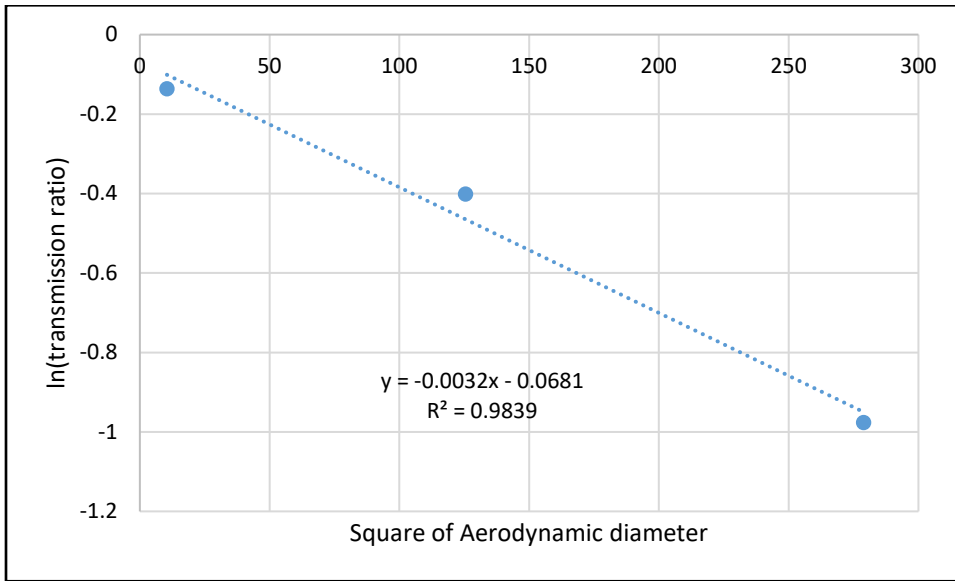


Figure 10. Plot of $\ln(P)$ transmission ratio versus (Da^2) aerodynamic diameter squared.

Based on the linear regression equation, the transmission ratio for different particle sizes can be calculated as:

$$P_j = \exp(-0.0032Da_j^2 - 0.0681) \quad (12)$$

Where: Da_j is the average aerodynamic particle diameter of APS sampling channel j ; P_j is the calculated transmission ratio.

Figure 11 shows the calculated transmission ratio for every APS sampling channel.

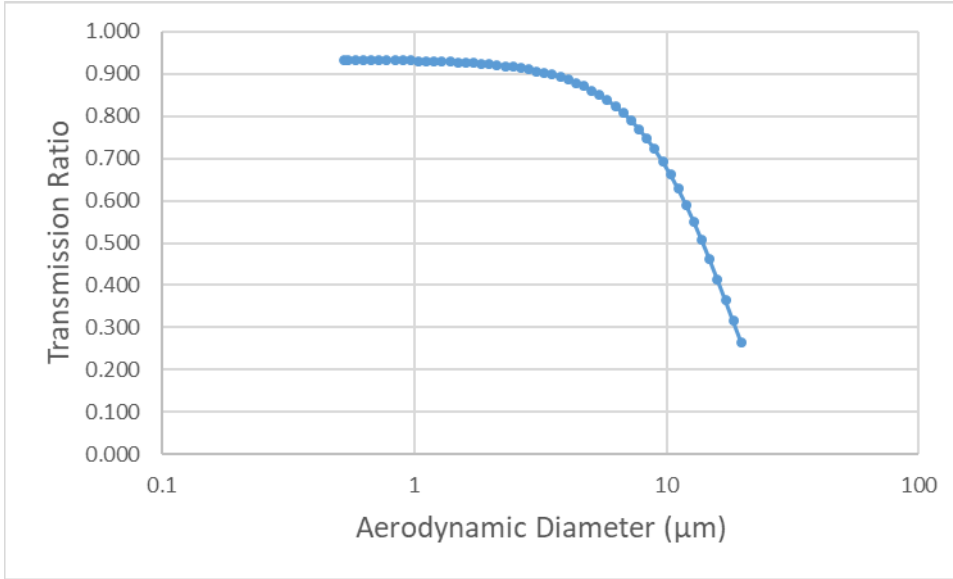


Figure 11. Calculated Transmission Ratio for APS sampling channels.

Respirable aerosol is defined (DOE 1994) as particles that can be transported through air and inhaled into human respiratory system and is commonly assumed to include particles 10 μm aerodynamic equivalent diameter and less. Therefore, CeO₂ particles that are detected in APS sampling channels 1 to 43 are respirable particles ($D_a \leq 10 \mu\text{m}$). Airborne aerosol is defined as particles that can be suspended in air and made available for airborne transport. Therefore, all CeO₂ particles that are detected in APS sampling channels (1- 52) are airborne particles (0.5 – 20 μm).

In a typical drop test, the RRFMC and wind tunnel are cleaned and prepared to reduce the background dust. Then the tested container is loaded in the desired drop orientation and the door of RRFMC is closed. The aerosol concentration inside the RRFMC and wind tunnel is monitored until it reaches the defined equilibrium background concentration (i.e. 10^{-5} mg/m^3). Then the tested container will be raised to the desired height and dropped by remote control. An increase of aerosol concentration will be detected by APS, whether there is a release of test powder or not, since even a clean dummy container will cause dust entrainment from the RRFMC. The aerosol concentration will be monitored until it returns to the background equilibrium value (Figure 12).

The respirable mass of aerosol released from a tested container, $mr(\text{mg})$, is calculated as:

$$mr = FR * Ts * \sum_{i=1}^t \left[\sum_{j=1}^{D_a=10} \left(\frac{C_{ij}}{P_j} \right) \right] \quad (13)$$

Where: C_{ij} is the measured aerosol concentration at $t=i$ in APS sampling channel j , mg/m^3 ;
 P_j is the transmission ration of APS sampling channel j , calculated by equation 12;
 FR is the RRFMC total air flow rate, measured at location #4, m^3/min
 Ts is the APS sampling interval, 1 min;
 $t=1$ is when the container is dropped;
 $t=t$ is when the measured concentration goes back to background concentration.

The airborne mass of aerosol released from a tested container, $ma(mg)$, is calculated as:

$$ma = FR * Ts * \sum_{i=1}^t \left[\sum_{j=1}^{D_a=20} \left(\frac{C_{ij}}{P_j} \right) \right] \quad (14)$$

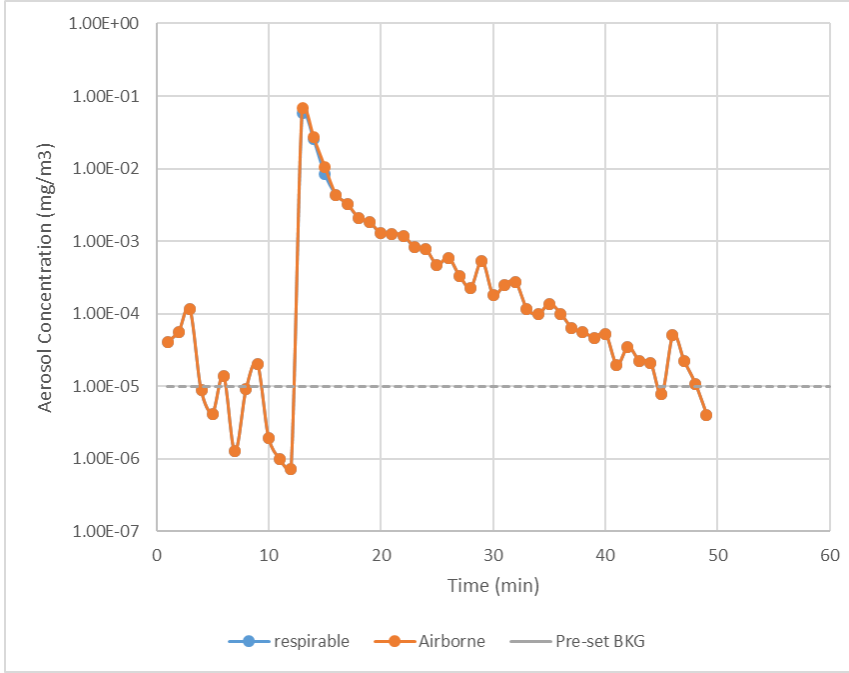


Figure 12. Aerosol concentration values in the RRFMC for a typical drop test.

The airborne source term (DOE 1994) is estimated by a five-component equation (Figure 13):

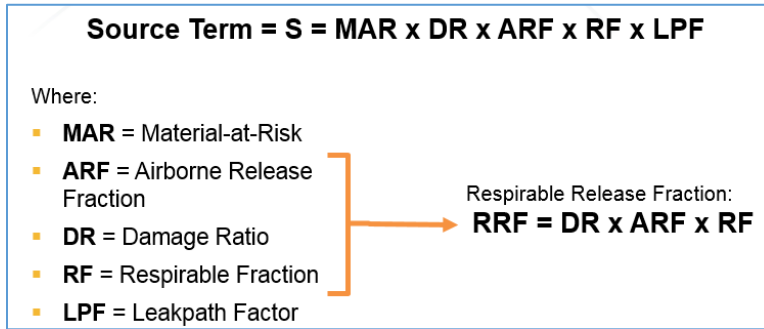


Figure 13. The five factor formula for source term estimation.

In the RRFMC, DR, ARF, RF, and RRF are calculated as:

$$DR = \frac{\Delta m}{MAR} \quad (15)$$

Where: Δm is the differential of initial to final container mass after drop test. Note if the container lid completely opens during a drop test, the DR is defined as 1.0 (unity).

$$ARF = \frac{ma}{MAR \cdot DR} \quad (16)$$

$$RF = \frac{mr}{ma} \quad (17)$$

$$RRF = DR \cdot ARF \cdot RF = \frac{mr}{MAR} \quad (18)$$

CONCLUSIONS

The aerosol transmission ratio of monodisperse oil droplets was measured in the Los Alamos RRFMC. Based on the experimental results, the transmission ratio for every APS sampling channels was calculated and respirable release mass, airborne release mass, RRF and ARF calculation formulas were derived.

REFERENCES

ANSI. Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks and Ducts of Nuclear Facilities. American National Standards Institute, Inc. ANSI/HPS Standard N13.1-2011. New York, USA. 2011.

Hinds WC. Aerosol Technology, Properties, Behavior and Measurement of Airborne Particles. John Wiley and Sons, USA, 2nd ed. 1999.

McFarland AR, Anand NK, Ortiz CA, Gupta R, Chandra S, and McManigle AP. A generic mixing system for achieving conditions suitable for single point representative effluent air sampling. Health Physics 76:17-26; 1999.

Moore ME, Fuehne DP and Schafer DP. Verifying a universal design for single point exhaust stack sampling. Unclassified report, LA-UR-10-04275. 2010.

Moore ME and McFarland AR. Design methodology for multiple inlet cyclone. Environmental Science & Technology 30:271-276. 1996.

Moore ME and Tao Y. Aerosol Physics Consideration for using Cerium Oxide CeO₂ as a Surrogate for Plutonium Oxide PuO₂ in Airborne Release Fraction measurements for Storage Container Investigations. Los Alamos Unrestricted Release LA-UR-17-21214. 2017.

Moore ME, Kennedy TJ, and Dimmerling PJ. Aerosol collection of the (Bladewerx Corporation) breathing zone monitor and portable workplace monitor. Health Physics 93:S165-S173, 2007.

ASME. Test uncertainty. The American Society of Mechanical Engineers. ASME PTC 19.1-2013. New York, NY, 2013.

U.S. Environmental Protection Agency. Procedures for testing performance characteristics of methods for PM₁₀. Washington, DC; U.S. Governmental Printing Office; Code of Federal regulations; 40 CFR Sec.53.40-44. 1997.

US DOE handbook DOE-HDBK-3010-94. Airborne release fractions/rates and respirable fractions for nonreactor nuclear facilities. US Department of Energy, 1994.